U.S. DEPARIMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES

DESIGNATED/ELECTED OFFICE (DO/EO/US)

CONCERNING A FILING UNDER 35 U.S.C. 371

29610/CDT091

US APPLICATION NO (IF KNOWN, SEE 37 CFR 10/009078

INTERNATIONAL APPLICATION NO. PCT/GB00/01991 INTERNATIONAL FILING DATE
1 June 2000

PRIORITY DATE CLAIMED
9 June 1999

TITLE OF INVENTION

METHOD OF PRODUCING ORGANIC LIGHT-EMISSIVE DEVICES

APPI	ICANT(S)	FOR	DO/	FO/I	IS

Paul R. BERGER and Stephen K. HEEKS

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Appl	icant l	nerewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:						
1.	$\boxtimes$	This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.						
2.		This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.						
3.	$\boxtimes$	This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include itens (5), (6) and (24) indicated below.						
4.	×	The US has been elected by the expiration of 19 months from the priority date (Article 31).						
5.	$\boxtimes$	A copy of the International Application as filed (35 U.S.C. 371 (c) (2))						
ı		a 🗵 is attached hereto (required only if not communicated by the International Burcau).						
ı		b. 🛮 has been communicated by the International Bureau.						
ı		c.   is not required, as the application was filed in the United States Receiving Office (RO/US).						
6.		An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).						
ı		a. Is attached hereto.						
		b. has been previously submitted under 35 U.S.C. 154(d)(4).						
7.	Z	Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))						
		a.   are attached hereto (required only if not communicated by the International Bureau).						
	-	b. have been communicated by the International Bureau.						
		c. have not been made; however, the time limit for making such amendments has NOT expired.						
		d. Me have not been made and will not be made.						
8.		An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).						
9.		An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).						
10.		An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).						
11.	$\bowtie$	A copy of the International Preliminary Examination Report (PCT/IPEA/409).						
12.	$\bowtie$	A copy of the International Search Report (PCT/ISA/210).						
I	tems 1	3 to 20 below concern document(s) or information included:						
13.	$\boxtimes$	An Information Disclosure Statement under 37 CFR 1.97 and 1.98.						
14.		An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3 31 is included						
15.	$\bowtie$	A FIRST preliminary amendment.						
16.		A SECOND or SUBSEQUENT preliminary amendment.						
17.		A substitute specification.						
18.		A change of power of attorney and/or address letter.						
19.		A computer-readable form of the sequence listing in accordance with PCT Rule 13ter 2 and 35 U.S.C. 1821 - 1.825.						
20.	0	A second copy of the published international application under 35 U.S.C. 454(d)(4).						
21.		A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).						
22.		Certificate of Mailing by Express Mail						
23.	$\boxtimes$	Other items or information						
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NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.													
SEND ALL CORRESPONDENCE TO:													
James P. Zeller													
Marshall, Gerstein & Borun						SIGNATURE							
233 S. Wacker Drive						James P. Zeller							
6300 Sears Tower Chicago, IL 60606						NAME							
(312) 474-6300					28,491								
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#### PATENT APPLICATION

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applic	cants: Berger et al.	)	"EXPRESS MAIL" mailing label No. EL564464835US.
Serial	No.: To be assigned	)	Date of Deposit: December 3, 2001
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For:	METHOD OF PRODUCING	)	MAIL POST OFFICE TO
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			Richard Zimmermann

#### PRELIMINARY AMENDMENT

Commissioner for Patents Box PCT Washington, DC 20231

Sir:

Please amend this application as follows.

#### IN THE SPECIFICATION:

Page 1 immediately following the title, please insert the following:

--This is the U.S. national phase of International Application No.

PCT/GB00/01991 filed June 1, 2000, the entire disclosure of which is incorporated herein by reference.--

JC10 Rec'd PCT/PTO 0 3 DEC 2001

#### IN THE ABSTRACT:

Please add an abstract as set forth on the attached sheet.

#### IN THE CLAIMS:

Please amend claims 2-19 and 22-26, as follows:

- (Amended) A method according to claim 1 comprising providing the holed layer by laminating a polymer layer with pre-formed holes to the surface of the substrate.
- 3. (Amended) A method according to claim 1 comprising providing the holed layer by providing a layer of a polymer on the substrate, and then forming the plurality of holes in the polymer layer.
- (Amended) A method according to claim 3 comprising forming the holes in the polymer layer by etching.
- (Amended) A method according to claim 4 comprising carrying out the etching using an oxygen-based plasma.
- (Amended) A method according to claim 4 comprising carrying out the etching by a plasma etching process.

- (Amended) A method according to claim 3 comprising providing the polymer layer by coating the substrate with the polymer.
- (Amended) A method according to claim 3 comprising providing the polymer layer by laminating a pre-formed polymer layer to the substrate.
- (Amended) A method according to claim 1 wherein the holed layer comprises an electrically insulating polymer.
- 11. (Amended) A method according to claim 1 wherein the thickness of the holed layer is in the range of 50nm to 100nm.
- 12. (Amended) A method for forming an optoelectronic device comprising the steps of:

providing a substrate having a patterned first electrode on a surface thereof;

providing a holed layer on the surface of the substrate having the first

patterned electrode, the holed layer being permanently attached to the substrate and
defining a first set of holes through which the patterned first electrode is exposed;

applying a first light-emissive material to the surface of the holed layer opposite the substrate;

displacing the first light-emissive material in fluid form across the surface of the holed layer so as to selectively deposit the material only in the first set of holes of the holed layer;

solidifying the first light-emissive material; and

forming a second electrode on the solidified first light-emissive material in the first set of holes such that charge carriers can move between the first light-emissive material and the second electrode.

- 13. (Amended) A method for forming an optoelectronic device according to claim 12 comprising providing the holed layer by laminating a polymer layer with pre-formed holes to the surface of the substrate.
- 14. (Amended) A method for forming an optoelectronic device according to claim 12 comprising providing the holed layer by providing a layer of a polymer on the surface of the substrate having the patterned first electrode, and then forming the first set of holes in the polymer layer.
- 15. (Amended) A method for forming an optoelectronic device according to claim 14 comprising forming the first set of holes by etching.
- 16. (Amended) A method for forming an optoelectronic device according to claim 12 wherein the patterned first electrode comprises a twodimensional array of discrete pixel electrodes.
- 17. (Amended) A method according to claim 16 wherein the first set of holes expose only some of the discrete pixel electrodes.

- 18. (Amended) A method according to claim 12 further comprising forming a second set of holes in the holed layer to expose discrete pixel electrodes which were not exposed by the first set of holes in the holed layer, depositing a layer of second light-emissive material in the second set of holes, and forming a third electrode on the layers of second light-emissive material such that charge carriers can move between the layers of second light-emissive material and the third electrode.
- 19. (Amended) A method according to claim 18 comprising additionally forming the second electrode over substantially the entire surface of the holed layer opposite the substrate, and forming the second set of holes by etching through the second electrode and the holed layer.
- 22. (Amended) A method according to claim 12 further comprising the step of forming a layer of a charge transport material in the first set of holes.
- 23. (Amended) A method according to claim 22 comprising depositing the layer of charge transport material in the first set of holes by applying the charge transport material to the surface of the holed layer and displacing the charge transport material in a fluid form across the surface of the holed layer whereby the charge transport material is deposited in the first set of holes.
- 24. (Amended) A method for forming an optoelectronic device according to any claim 12 comprising depositing the second electrode without first removing the holed layer.

- 25. (Amended) A method for forming an optoelectronic device according to claim 20 comprising forming the second, third and fourth electrodes without removing the holed layer.
- 26. (Amended) An optoelectronic device produced by the method of claim 12.

Please add new claims 27 and 28, as follows:

- 27. A method according to claim 5 comprising carrying out the etching by a plasma etching process.
  - 28. An optoelectronic device produced by the method of claim 1.

#### REMARKS

By the foregoing amendments to the specification, a cross-reference to the parent international application has been provided. The claims have been amended to better conform to U.S. practice and to omit multiple dependencies.

The filing fee has been calculated based on the claims as amended above. No new matter has been added.

Respectfully submitted,

MARSHALL, GERSTEIN & BORUN

December 3, 2001

James P. Zeller

6300 Sears Tower 233 South Wacker Drive Chicago, Illinois 60606-6402 (312) 474-6300

#### VERSION WITH MARKINGS TO SHOW CHANGES MADE

#### IN THE SPECIFICATION:

Page 1 immediately following the title, please insert the following:

--This is the U.S. national phase of International Application No.

PCT/GB00/01991 filed June 1, 2000, the entire disclosure of which is incorporated herein by reference.--

Please amend claims 2-19 and 22-26, as follows:

- (Amended) A method according to claim 1 [wherein] comprising
   providing the holed layer [is provided] by laminating a polymer layer with pre-formed holes to the surface of the substrate.
- 3. (Amended) A method according to claim 1 [wherein] comprising providing the holed layer [is provided] by providing a layer of a polymer on the substrate, and then forming the plurality of holes in the polymer layer.
- 4. (Amended) A method according to claim 3 [wherein] comprising forming the [holes are formed] holes in the polymer layer by etching.
- 5. (Amended) A method according to claim 4 [wherein] comprising carrying out the etching [is carried out] using an oxygen-based plasma.

- (Amended) A method according to claim 4 [or 5 wherein]

  <u>comprising carrying out</u> the etching [is carried out] by a plasma etching process.
- 7. (Amended) A method according to claim 3 [wherein] comprising providing the polymer layer [is provided] by coating the substrate with the polymer.
- 8. (Amended) A method according to claim 3 [wherein] <u>comprising</u> <u>providing</u> the polymer layer [is provided] by laminating a pre-formed polymer layer to the substrate.
- (Amended) A method according to [any preceding] claim 1 wherein the holed layer comprises an electrically insulating polymer.
- 11. (Amended) A method according to [any preceding] claim  $\underline{1}$  wherein the thickness of the holed layer is in the range of  $50 \underline{nm}$  to  $100 \underline{nm}$ .
- 12. (Amended) A method for forming an optoelectronic device comprising the steps of:

providing a substrate having a patterned first electrode on a surface thereof;

providing a holed layer on the surface of the substrate having the first

patterned electrode, the holed layer being permanently attached to the substrate and
defining a first set of holes through which the patterned first electrode is exposed;

applying a first light-emissive material to the surface of the holed layer opposite the substrate[, and];

displacing the first light-emissive material in fluid form across the surface of the holed layer so as to selectively deposit the material only in the first set of holes of the holed layer;

solidifying the first light-emissive material; and

forming a second electrode on the solidified first light-emissive material in the first set of holes such that charge carriers can move between the first light-emissive material and the second electrode.

- 13. (Amended) A method for forming an optoelectronic device according to claim 12 [wherein] comprising providing the holed layer [is provided] by laminating a polymer layer with pre-formed holes to the surface of the substrate.
- 14. (Amended) A method for forming an optoelectronic device according to claim 12 [wherein] <u>comprising providing</u> the holed layer [is provided] by providing a layer of a polymer on the surface of the substrate having the patterned first electrode, and then forming the first set of holes in the polymer layer.
- 15. (Amended) A method for forming an optoelectronic device according to claim 14 [wherein] comprising forming the first set of holes [are formed] by etching.
- 16. (Amended) A method for forming an optoelectronic device according to [any one of claims] <a href="claim">claim</a> 12 [to 15] wherein the patterned first electrode comprises a two-dimensional array of discrete pixel electrodes.

- 17. (Amended) A method according to claim 16 wherein the first set of holes [only] expose only some of the discrete pixel electrodes.
- 18. (Amended) A method according to claim 12 further comprising forming a second set of holes in the holed layer to expose discrete pixel electrodes which were not exposed by the first set of holes in the holed layer, depositing a layer of second light-emissive material in the second set of holes, and forming a third electrode on the layers of second light-emissive material such that charge carriers can move between the layers of second light-emissive material and the third electrode.
- 19. (Amended) A method according to claim 18 [wherein] <u>comprising</u> additionally forming the second electrode [is additionally formed] over substantially the entire surface of the holed layer opposite the substrate, and <u>forming</u> the second set of holes [is formed] by etching through the second electrode and the holed layer.
- 22. (Amended) A method according to [any of claims] <u>claim</u> 12 [to 21] further comprising the step of forming a layer of a charge transport material in the first set of holes.
- 23. (Amended) A method according to claim 22 [wherein] <u>comprising</u> <u>depositing</u> the layer of charge transport material [is deposited] in the first set of holes by applying the charge transport material to the surface of the holed layer and displacing the charge transport material in a fluid form across the surface of the holed layer whereby [it] the charge transport material is deposited in the first set of holes.

- 24. (Amended) A method for forming an optoelectronic device according to any [one of claims] claim 12 [to 23 wherein] comprising depositing the second electrode [is deposited] without first removing the holed layer.
- 25. (Amended) A method for forming an optoelectronic device according to claim 20 [wherein] comprising forming the second, third and fourth electrodes [are formed] without removing the holed layer.
- 26. (Amended) An optoelectronic device produced by the method of [any of claims 14 to 25] claim 12.

Please add new claims 27 and 28, as follows:

- A method according to claim 5 comprising carrying out the etching by a plasma etching process.
  - 28. An optoelectronic device produced by the method of claim 1.

#### ABSTRACT

A method for forming a patterned layer of a light-emissive material on a substrate, comprising the steps of providing a holed layer on the surface of the substrate, the holed layer being permanently attached to the substrate and defining a plurality of holes through which the underlying substrate is exposed, and applying a light-emissive material to the surface of the holed layer opposite the substrate and displacing the light-emissive material in fluid form across the surface of the holed layer so as to selectively deposit the material only in the holes of the holed layer.

# 1.07.07.07.28 1:07.009.07.8 PTO/PCT Rec'd, 0,3 DEC 2001

#### WO 00/76008

#### METHOD OF PRODUCING ORGANIC LIGHT-EMISSIVE DEVICES

The present invention relates to a method for producing optoelectronic devices, in particular, organic light-emissive devices (OLEDs).

Organic light-emissive devices typically comprise an organic light-emissive region sandwiched between two electrodes such that charge carriers can move between the organic light-emissive region and the two electrodes.

In many applications such as active matrix displays, at least one of the electrodes and the light-emissive region is required to be patterned into an ordered array of discrete pixels whereby each pixel can be addressed separately. Full colour active matrix displays require a light-emissive region comprising discrete pixels of three different light-emissive materials which are respectively capable of emitting light in the three primary colour regions.

One method of providing a patterned organic light-emissive region involves the use of ink-jet printing. However, the degree of pixel density which can be achieved by ink-jet printing is severely limited. Furthermore, the droplets formed by ink-jet printing are hemispherically shaped leading to non-uniform illumination within each pixel. In addition, it is an expensive process particularly when used to produce large area displays.

According to a first aspect of the present invention, there is provided a method for forming a patterned layer of a light-emissive material on a substrate, comprising the steps of providing a holed layer on the surface of the substrate, the holed layer being permanently attached to the substrate and defining a plurality of holes through which the underlying

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substrate is exposed; and applying a light-emissive material to the surface of the holed layer opposite the substrate and displacing the light-emissive material in fluid form across the surface of the holed layer so as to selectively deposit the material only in the holes of the holed layer.

The method of the present invention can be used to form any patterned layer such as a series of parallel rows or columns, a two-dimensional array of discrete pixels, or a layer in the shape of a legend comprising, for example, one or more letters or numerals or other shapes.

According to the present invention, the light-emissive material is displaced across the surface of the holed layer such that it is selectively deposited in the holes of the holed layer, i.e. such that it is deposited in the holes of the holed layer without substantially any deposition of the light-emissive material on the surface of the holed layer. Furthermore, the holed layer is permanently adhered to the substrate, i.e. the holed layer is not intended to be subsequently removed from the substrate. Indeed, the holed layer may be integral with the substrate.

The light-emissive material is preferably an organic light-emissive material. The term organic light-emissive material includes precursors of organic light-emissive materials, the precursors not necessarily being light-emissive themselves. For example, the organic light-emissive material may be a semi-conductive conjugated polymer or a precursor of a semi-conductive conjugated polymer.

The holed layer is preferably made of a polymer which is electrically non-conductive, and does not chemically react with the light-emissive material in a way which renders the light-

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emissive material non light-emissive. The holed layer may be opaque or transparent.

The holed layer is preferably made of a relatively benign polymer such as a fluorinated polymer. Examples of suitable fluorinated polymers include amorphous polytetrafluoroethylene (PTFE) or an amorphous ethylene-tetrafluoroethylene copolymer (ETFE), with amorphous PTFE being particularly suitable. The use of fluorinated polymers is advantageous because they do not react and swell upon contact with the standard solvents such as xylene and water which are typically used for forming solutions of organic light-emissive materials.

The holed layer is preferably formed by providing a layer of a polymer on the substrate, and then forming a plurality of holes in the polymer layer by etching. The etching is preferably carried out by a reactive ion etching process using an oxygen-based plasma. The polymer layer may be provided on the substrate by deposition using a standard technique such as spin-coating or blade-coating, or by laminating a pre-formed polymer layer to the substrate.

Alternatively, a polymer layer with pre-formed holes could be prepared in advance and then laminated to the substrate.

The thickness of the holed layer will clearly depend on the desired thickness of the patterned layer of light-emissive material, since its thickness will determine the thickness of the patterned layer of light-emissive material. For a typical organic light-emissive device, the fluorinated polymer layer is deposited to a thickness in the range of 50 to 100 nm.

The light-emissive material is preferably applied to the surface of the holed layer as a solution thereof in a suitable solvent.

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Insoluble materials can be applied, for example, as a suspension thereof in a suitable liquid medium or in molten form.

According to a second aspect of the present invention, there is provided a method for forming an optoelectronic device comprising the steps of providing a substrate having a patterned first electrode on a surface thereof, providing a holed layer on the surface of the substrate having the first patterned electrode, the holed layer being permanently attached to the substrate and defining a first set of holes through which the pattered first electrode is exposed, applying a first lightemissive material to the surface of the holed layer opposite the substrate, and displacing the first light-emissive material in fluid form across the surface of the holed layer so as to selectively deposit the material only in the first set of holes the holed layer, solidifying the first light-emissive material, and forming a second electrode on the solidified first light-emissive material in the first set of holes such that charge carriers can move between the first light-emissive material and the second electrode.

The light-emissive material can be deposited directly on the underlying patterned first electrode. Alternatively, intermediate layers such as charge transport lavers additional light-emissive layers can be provided between the patterned first electrode and the light-emissive material provided that they allow the movement of charge carriers between the patterned first electrode and the light-emissive material. In a preferred embodiment, a layer of a hole-injection material is provided between the patterned first electrode and the lightemissive layer to promote the injection of holes from the patterned first electrode into the light-emissive material.

Likewise, the second electrode can be formed directly on the layer of light-emissive material, or intermediate layers such as

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charge transport layers or additional layers of light-emissive material can be interposed between the layer of light-emissive material and the second electrode, provided that they allow the movement of charge carriers between the second electrode and the layer of light-emissive material.

The light-emissive material is preferably an organic light-emissive material such as a semi-conductive conjugated polymer. The term conjugated polymer includes polymers and oligomers which are conjugated along the entire length thereof, and polymers and oligomers which comprise conjugated segments separated by non-conjugated segments.

The holed layer is preferably formed by depositing a layer of a polymer on the substrate and then forming the plurality of holes in the polymer layer after deposition of the holed layer. The plurality of holes are preferably formed by etching of the polymer layer, such as anisotropic reactive ion etching.

A patterned array of one or more holes of the desired shape may be achieved by a photolithographic technique. Such a technique involves forming a layer of a photoresist on the surface of the polymer layer; exposing the whole area of the photoresist (for example, using a shadow mask) to radiation; removing the portions of the photoresist layer by the use of a suitable developer to leave a photoresist layer having one or more holes of the desired shape formed therein which expose the portions of the underlying polymer layer in which holes are to be formed. Subsequent exposure to an etching plasma will only etch the portions of the polymer layer left exposed by the holes in the photoresist layer.

The etching is preferably carried out using a plasma which not only serves to etch the material of the holed layer but also

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serves to treat and prepare the exposed surface of the patterned first electrode for deposition of subsequent layers.

Embodiments of the present invention will be described hereunder, by way of example only, with reference to the accompanying drawings, in which:-

Figures 1(a) to 1(h) are schematical cross-sectional views of a single colour organic light-emissive device at various stages of its production according to a first embodiment of the present invention.

Figures 2(a) to 2(h) are schematical cross-sectional views of a full colour organic light-emissive device at various stages of its production according to a second embodiment of the present invention.

Figure 3 shows an organic light-emissive device incorporating a resonant cavity structure and a light-dispersal structure.

First, with reference to Figures 1(a) to 1(h), a method of producing an organic light-emissive device according to a first embodiment of the present invention will be described.

A layer of a fluorinated polymer 6 such as amorphous polytetrafluoroethylene (PTFE) is formed over the surface of a substrate comprising a glass base 2 having a two-dimensional ordered array of anode pixels 4 made of indium tin oxide (ITO) formed on a surface thereof. In this embodiment, the substrate is made of glass, but it can also be made of other materials such as plastics eg. PMMA. Wiring (which is not shown in the Figures) is connected from the periphery of the glass base to each anode pixel so that each anode pixel can be separately addressed in the final device.

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The two-dimensional array of anode pixels 4 can be formed on a glass base 2 according to a conventional method such as sputtering using a shadow mask. The layer of fluorinated polymer is formed by a conventional coating method such as spin-coating using a solution of the fluorinated polymer in a suitable organic solvent and then drying.

The thickness of the layer of the fluorinated polymer layer will determine the thickness of the organic light-emissive region in the final device. For a typical organic light-emissive device, the fluorinated polymer layer is deposited to a thickness in the range of 50 to 100nm.

Holes 8 are then created in the fluorinated polymer layer 6 to expose portions of the underlying anode pixels 4. The holes 8 are created by the following process. The surface of the fluorinated polymer layer is provided with an appropriately patterned photopolymer layer such as a photoresist on its upper surface by a standard photolithographical technique. The use of projection lithography to produce the patterned photopolymer layer can give affordable and high throughput processing.

Etching of the regions of the fluorinated polymer layer 6 left exposed by the photoresist is then carried out by a plasma etching process using an oxygen-based chemistry to form vertical holes in the fluorinated polymer layer 6 which extend down to the surface of the anode pixels 8. Each anode pixel 4 serves as an etch-stop. Once etching is completed the photoresist is stripped from the surface of the holed layer 6. The oxygen plasma also serves to treat the surface of the ITO anode pixels 4 for subsequent deposition of organic layers.

Although an oxygen plasma is used in this embodiment, other gases such as freons and sulphur hexafluoride can be used to carry out the etching process, with or without dilutions of

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oxygen. Alternatively, argon could be used where selectivity is not a requirement.

The exposed surface of the ITO anode pixels is then cleaned. Once the surface is cleaned, another short oxygen plasma treatment may be required to recover the treated ITO surface.

Next, a solution 12 of polystyrene sulphonic acid doped polyethylene dioxythiophene ("PEDOT-PSS") in water is applied to the surface of the holed polymer layer 6 and is wiped across the surface of the holed polymer layer 6 using a push rod 10, whereby the PEDOT-PSS solution 12 becomes deposited in the holes 8 to completely fill the holes 8 without substantially any deposition of the PEDOT-PSS solution on the top surface of the holed layer 6. The PEDOT-PSS solution 12 in the holes 8 is then dried to leave PEDOT-PSS pixels 14 in the holes 8. The PEDOT-PSS pixels only partially fill the holes 8 as a consequence of the reduction in volume associated with the evaporation of the agueous solvent.

The thickness of the PEDOT-PSS pixels can be controlled by adjusting the concentration of the PEDOT-PSS solution. The thickness of the PEDOT-PSS pixels may be reduced by using a more dilute solution, and may be increased by using a more concentrated solution.

The use of a fluorinated polymer for the holed layer 6 is advantageous in this embodiment because it does not react and swell upon contact with the aqueous solvent.

The PEDOT-PSS provides a hole-injection layer which promotes injection of positive charge carriers into the light-emissive material to be subsequently deposited. Other materials which provide this hole-injecting function can also be used such as poly(2,7-(9,9-di-n-octylfluorene)-(1,4-phenylene-(4-

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imino(benzoic acid))-1,4-phenylene-(4-imino(benzoic acid))-1,4phenylene)) ("BFA"), polyaniline and poly(p-phenylenevinylene)
("PPV").

Next, a solution 16 of an organic light-emissive material such as a semi-conductive conjugated polymer is applied to the surface of the holed layer 6 and is wiped across the surface of the holed layer 6 using a push rod 10 whereby it is selectively deposited in the holes 8 in the holed layer 6 on top of the PEDOT-PSS pixels 14, again to completely fill the holes without substantially any depostion of the organic light-emissive material on the top surface of the holed layer 6. The solution of the organic light-emissive material 16 left deposited in the holes 8 of the holed layer 6 is then dried to leave each hole containing an underlying layer 14 of PEDOT-PSS and an overlying layer of the organic light-emissive material. The selective deposition technique described above constrains the amount of material in fluid form deposited at each location to the available volume of the corresponding hole in the holed layer. This in turn is determined by the dimensions of the hole, which can be accurately controlled: the hole depth by the thickness of the holed layer 6, the dimensions in the plane of the layer by the photolithographic etching process. The above technique therefore allows the amount of fluid deposited at each location to be accurately controlled and, when used in combination with a carefully controlled concentration, allows thickness of the resulting solid film to be accurately controlled. As is known, film thickness can be critical for the efficiency of organic electroluminescent devices.

The organic light-emissive material may comprise one or more individual organic materials, suitably polymers, preferably fully or partially conjugated polymers. Suitable materials include one or more of the following in any combination: poly(p-phenylenevinylene) ("PPV"), poly(2-methoxy-5(2'-

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ethyl)hexyloxyphenylenevinylene) ("MEH-PPV"), one or more PPVderivatives (e.q. di-alkoxv or di-alkvl derivatives). polyfluorenes and/or co-polymers incorporating polyfluorene segments, PPVs and related co-polymers, poly(2,7-(9,9-di-noctylfluorene)-(1,4-phenylene-((4-secbutylphenyl)imino)-1,4phenylene)) ("TFB"), poly(2,7-(9,9-di-n-octylfluorene) - (1,4phenylene-((4-methylphenyl)imino)-1,4-phenylene-((4 methylphenyl)imino) - 1,4-phenylene)) ("PFM"), poly(2,7 - (9,9 di-n-octylfluorene) - (1,4-phenylene-((4-methoxyphenyl)imino)-1,4-phenvlene-((4-methoxyphenyl)imino)-1,4-phenylene)) ("PFMO"), poly (2,7-(9,9-di-n-octylfluorene) ("F8") or (2,7-(9,9-di-n-octylfluorene)-3,6-Benzothiadiazole) ("F8BT"). Alternative materials include small molecule materials such as Ala3.

A typical solvent for forming solutions of the above materials is xylene. Other suitable solvents are THF, trichloromethane, toluene and other aromatic solvents. When these are used as the solvent, the use of a fluorinated polymer for the holed layer is advantageous because it does not swell upon contact with them.

A cathode 20 is then deposited over the surface of the lightemissive pixels 18 and the surface of the holed layer 6. The cathode preferably has a double layer construction with a thin underlying layer of a reactive metal such as calcium deposited on top of the light-emissive pixels 18 and a thick layer of a non-reactive metal such as aluminium deposited on top of the thin layer of reactive metal. The cathode is preferably deposited by evaporation to reduce to a minimum any damage to the underlying organic layers.

The matrix of polymer material surrounding each organic pixel and each anode pixel remains in the final device, and further serves to both electrically insulate the organic pixels and anode pixels from the influence of adjacent organic pixels and

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anode pixels. It also serves to physically protect and give structural support to the organic pixels and anode pixels.

Next, with reference to Figures 2(a) to 2(h), a method of producing a full colour organic light-emitting device according to a second embodiment of the present invention will be described.

A layer 54 of a fluorinated polymer is formed on a substrate comprising a glass base 50 having a two-dimensional array of anode pixels 52 made of ITO deposited thereon. The fluorinated polymer layer 54 may be formed, for example, by the method described for the first embodiment.

A first set of holes 56 are then formed at selected points of the fluorinated polymer layer 54 in the same way as in the first embodiment, to expose only some of the anode pixels 52. For example, the two dimensional array of anode pixels may comprise an array of groups of three associated pixels, with the first set of holes 56 being formed to expose only a single one of the three anode pixels in each group.

A layer of an organic hole-injection material and a layer of a first organic light-emissive material are then deposited in each hole 56 in the same manner as in the first embodiment to create an array of organic pixels 58. A cathode 60 is then formed over the whole surface of the holed layer 52 and the organic pixels 58. As in the first embodiment, the cathode preferably has a double layer construction with an underlying first thin layer of a reactive metal such as calcium and an overlying thick layer of a non-reactive metal such as aluminium.

The cathode metallization serves to protect the organic pixels 58 embedded into the fluorinated polymer from subsequent processing steps.

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Next, a second set of holes 62 is formed to expose some of the anode pixels 52 which were not exposed in the previous holeforming step. For example, in the array described above, the second set of holes could be formed to only expose a second one of the three anode pixels in each group. This second set of holes 62 can be formed in the same manner as the first set of holes, except that different plasma chemistries may be one to punch through the cathode 60 and one to etch through the fluorinated polymer laver 54. Fluorocarbons. hexafluoride, argon, methane/hydrogen and chlorine chemistries such as boron trichloride are examples of materials which can be used to punch through the cathode. The etching through of the cathode can be observed by optical absorption in the plasma chamber used. The etching of the fluorinated polymer layer 54 is carried out using an oxygen-based plasma as in the first embodiment.

A second set of organic pixels 64 is then formed in the second set of holes 62 in the same manner as in the first embodiment. Although, not shown in the Figures, each of the second set of organic pixels 64 also comprises an underlying layer of PEDOT-PSS (as a hole injection layer) and an overlying layer of an organic light-emissive organic material which is capable of emitting light of a primary colour different to that of the light-emissive material deposited in the first set of holes 56.

Another cathode 66 is then deposited over the second set of organic pixels 64 to protect the second set of organic pixels 58 from subsequent processing steps.

Next, a third set of holes 68 is formed through the cathodes 60,66 and the fluorinated polymer layer 54 to expose the remaining anode pixels which were not exposed in the previous first and second hole forming steps. For example, in the array

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described above, the third set of holes could be formed to only expose the third and final anode pixel of each group of three associated anode pixels. The third set of holes 68 can be formed in the same manner as the second set of holes 62. A third set of organic pixels 70 is then deposited in the third set of holes 68 in the same manner as in the first embodiment. Although not shown in the Figures, the third set of organic pixels 68 also each comprise an underlying layer of PEDOT-PSS as a hole-injection layer and an overlying layer of an organic light-emissive material which is capable of emitting light of a primary colour different from the light-emissive materials deposited in the first and second sets of holes 56, 62.

In both the above-described embodiments, the fact that the shape and size of the organic pixels are defined by holes which are created by etching means that pixels of uniform shapes and of very small dimensions (of the order of a number of microns) can be formed because of the high precision of the etching process, and the pixel density can therefore be increased significantly compared to arrays of organic pixels produced by conventional methods such as ink-jet printing.

Furthermore, this method can be used to produce pixels having a uniform rectangular shape i.e. having a depth greater than their length or width. This gives good colour uniformity across the individual pixel area.

Furthermore, the above-described method of deposition of the organic materials is an extremely cost-effective deposition method which is particularly significant in the production of large area displays.

A final advantage of the above-described method is its ability to accurately meter the amount of fluid material deposited and

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thereby control the thickness of the resultant dried film, this being important for device efficiency.

Figure 3 shows a light-emitting device comprising: a pair of electrodes; a light emitting region located between the electrodes and comprising light-emitting organic material; a first reflective layer and a second reflective layer, the second reflective layer being partially transmissive and the first and second reflective layers being located on either side of the light-emitting region to define a resonant cavity structure about the light-emitting region; and a light-dispersal structure outside the cavity for dispersing light emitted from the cavity through the second reflective layer.

The OLED of figure 3 includes a region 101 of light emitting material. An anode electrode 102 and a cathode electrode 103 are located on either side of the light-emitting material. A resonant cavity reflector structure is defined about the light-emitting region 101 by a reflective layer 104 and a partially reflective and partially light-transmissive layer 105. The resonant structure is capable of enhancing emission from the light-emitting region and/or narrowing the emission spectrum from the region. On the outside of the partially light-transmissive layer is a structure 106 that is capable of dispersing the light emitted from the cavity so as to render the far-field emission from the overall device more Lambertian.

The resonant structure of the device constitutes a planar microcavity. The resonance wavelengths,  $\lambda_{\text{Re.}}$ , of the cavity are determined by the effective length,  $L_{\text{eff}}$ , of the cavity. The effective length of the cavity is a function of the refractive index and the thickness of each of the layers in the cavity and the phase change on reflection of light from the mirrors. If the refractive index of the  $\kappa$ th layer of the cavity is  $n_k$  and

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the thickness of the  $\kappa$ th layer of material is  $d_k$  then the effective length of the cavity is given by:

$$L_{eff} = L_{phave\_change} + \sum_{k} n_k \cdot d_k$$

and the resonance wavelengths are given by:

$$\lambda_{\text{Re }s} = \frac{2L_{\text{eff}}}{q}$$

where q is an integer.

The electroluminescent device of figure 3 is formed on a glass substrate 109 which is coated with an anode electrode layer 102 of indium-tin oxide (ITO). Such ITO-coasted glass substrates are commercially available. The glass sheet could be a sheet of sodalime or borsilicate glass of a thickness of, for instance 1mm. Instead of glass other materials such as Perspex could be used. The thickness of the ITO coating is suitably around 150nm and the ITO suitably has a sheet resistance of between 10 and  $30\Omega/$ , preferably around  $15\Omega/$ . Instead of ITO other materials, preferably of relatively high work function, such as tin oxide (TO) could be used.

A distributed Bragg reflector (DBR) consists of a stack of regularly alternating higher—and lover-refractive index dielectrics (light transmissive materials) fabricated to fulfil the Bragg condition for reflection at particular wavelengths. This occurs when the optical path of the periodicity in the dielectric stack corresponds to half a wavelength, and the reflectivity is further optimised when the DBR stack obeys the following condition:

 $\frac{1}{2}\lambda = n_1d_1 + n_2d_2$ 

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where  $n_1$ ,  $n_2$  are the respective refractive indices;  $d_1$ ,  $d_2$  are the corresponding component film thicknesses in the DBR; and  $\lambda$  is the desired reflection wavelength.

The reflective layer 104 is formed over the ITO layer 102. The reflective layer 104 is formed as a distributed Bragg reflector structure formed of alternating layers of conductive semiconductive materials such as partially doped PPV or other conjugated polymers (see our co-pending UK patent application number 9907802.4, the contents of which are incorporated herein by reference) or GaAs and AlGaAs. The layer thicknesses are chosen so that the structure is capable of reflecting light at a selected emission frequency of the emissive region. Instead of the reflective layer 104 being formed between the electrode 102 and the light-emitting layer 101 the electrode 102 could be formed between the light-emitting layer 101 and the reflective In that case the reflective layer would not have to be electrically conductive, but the electrode 102 should be lighttransmissive, as is ITO, for instance. Another alternative is for the reflective structure to be formed over only parts of the ITO layer - in that case the reflector could be electrically insulating but conduction from the ITO to the emissive layer is possible in the regions not covered by the reflective layer. Suitable materials for an electrically insulating DBR include Si and  $SiO_2$ . A further alternative is to form the ITO layer 102 to a thickness of a quarter of the (or a desired) emission wavelength of the light-emitting material, so it acts as at partial DBR structure - the separate reflective structure could then be omitted.

In the device of figure 3 a charge transport layer 107 is formed over the reflective layer 104. The charge transport layer improves hole transport between the anode and/or reflective structure 104 and the light-emitting region 101. The charge

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transport layer is formed of PEDOT:PSS. The charge transport layer could be formed from a solution containing PEDOT:PSS with a ratio of PEDOT to PSS of around 1 to 5. The hole transport layer could be spin-coated from solution and then baked e.g. at 200°C for 1 hour in a nitrogen environment. The thickness of the transport layer is suitably around 50nm but the thickness may be chosen so as to set the width of the resonant cavity to a desired value whilst allowing the thicknesses of the other layers of the device to be optimised for other factors. Instead of PEDOT:PSS other conductive materials such a polyaniline could be used for the charge transport layer 107. Alternatively the charge transport layer could be omitted and the thickness of other layers of the device could be chosen so as to fix the width of the cavity.

Then the light emitting layer 101 is deposited. The light-emitting layer could be formed of any suitable organic light-emitting material, but polymer materials are preferred. For example, an electroluminescent layer comprising 20% TFB in 5BTF8 could be coated over the hole transport layer by spin-coating typically to a thickness of 90nm. The term 5BTF8 refers to poly (2,7-(9,9-di-n-octylfluorene) ("F8") doped with 5% poly-(2,7-(9,9-di-n-octylfluorene)-3,6-benzothiadiazole) ("F8BT").

The cathode electrode layer 103 is formed over the light-emitting layer 101. The cathode electrode layer is formed of LiF with a thickness of 0.5 to 10nm, formed by evaporation of LiF. Such a layer is light-transmissive, so that light can pass through it to leave the device. In the device of figure 3 the reflector 104 is fully reflective and emission is from the upper surface of the device as illustrated. Therefore the cathode is formed of a light-transmissive material. If the reflector 104 were partially reflective and emission from the lower surface of the device, the cathode could be opaque and formed of e.g. thin layer of Ca covered with a protective layer of Al. It is

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preferred that the material of the cathode electrode, at least at the surface facing the light-emitting material, has a relatively low work function.

In this embodiment a further DBR structure 105 is formed over the cathode electrode. The structure 105 is formed as structure 104, but with fewer layers so that the structure 105 fully reflective but is partially reflective partially light transmissive. This allows some leakage from the cavity structure so that light can leave the device. A suitable form for the structure 105 is again alternating layers of differently doped conjugated polymer(s), alternating layers of GaAs/AlGaAs or alternating lavers of Si/SiO2. Since structure 105 is not located between the electrodes it need not be electrically conductive or semiconductive but could be formed of alternating layers of insulating materials. Alternatively, the cathode layer 103 could be metallised so that is partially reflective and partially light-transmissive, so that there is no need for a separate reflective layer.

The light dispersal structure 106 is formed over the upper reflector of the cavity -i.e. outside (preferably immediately outside) the cavity in the viewing direction. In the embodiment of figure 3 the light-dispersal structure is formed by coating the device with a layer of light-transmissive polymer material containing a dispersal; of light-transmissive particles 108. The particles have a different refractive index from the polymer matrix, so light leaving the device is scattered as it passed through the structure. The particles could be fragments or spheres of glass or latex or nanoparticles of a material such as TiO<sub>2</sub> (see our co-pending UK patent application number 9815270.5, the contents of which are incorporated herein by reference). Suitable materials for the matrix include fluorinated polymers, polyimide, polyethylene or a photoresist material. The diameter

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of the particles could suitably be 0.1 to 0.5 times the selected emission wavelength of the device.

It may be preferred for the particles to have a size distribution across all or part of that range or outside it in order to lessen any wavelength dependence of the scattering-this may be useful if the device is to be a full-colour display. The thickness of the layer of matrix could be 10-5000nm and the loading of the particles 10-60% by volume. The structure could be formed by spin-coating, spray coating, evaporation or lamination. The materials of the particles and the matrix should be chosen so that they have different refractive indices (so that scattering can occur in the structure) and also so that the refractive index of the matrix does not cause undesirable reflections at its interface with the adjacent underlaying layer.

Instead of containing solid particles, the matrix of the light dispersal structure could be formed so as to include bubbles for pockets of air that could similarly perform scattering of the emitted light.

Finally, contacts are made to the anode and cathode electrodes to allow the device to be driven, and the device is encapsulated, for example in epoxy, for environmental protection.

When the device is in use emitted light is scattered by the structure 106, which renders the far-field emission from the device more Lambertain. After scattering, the light is emitted in a range of directions from each point over the device's area. The range of directions is preferably greater than 90° and may preferably approach 180°.

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The structure 106 could be formed outside, rather than inside the encapsulant. Forming the structure 106 atop the encapsulant may help to reduce the possibility of waveguiding in the device. If the encapsulant itself provides the scattering structure then it is especially preferable for the scattering to be by means of bubbles or voids (e.g. containing air or inert gas) in the encapsulant layer.

The structure 106 could be formed in many other ways. For example, the structure could be formed by a layer (for example of a polymer material such as fluorinated polymers, polyimide, polyethylene or a photoresist material) having surface relief on its upper surface (the surface further from the light-emitting region) so that it causes scattering of emitted light there. The surface relief could be formed by etching (e.g. plasma or wet etching) into the surface, either randomly or in a predetermined design, by stamping or scratching or in another The predetermined design could be chosen to promote Lambertian output. The upper surface could be uneven or rough, and could include a pattern of scratches, grooves, ridges or other surface irregularity which could be formed by scraping or The scale of the roughening is suitably in the size range that will cause efficient scattering of the light emitted from the cavity -e.g. in the range from 0.1 to 0.5 times the emission wavelength of the device.

In the embodiment of figure 3 the reflector 105 is partially reflective and the electrode 103 is light transmissive. In other devices the reflector 104 could be partially reflective and the electrode 102 light transmissive.

The electrodes could be reversed, so that the first electrode to be deposited is the cathode and the upper electrode the anode.

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A charge transport layer could be provided between the emissive material the cathode to improve electron transport into the light-emitting region.

Either or both of the electrodes may be patterned so to allow individual regions of the display to be selectively addressed.

The particles could be opaque, provided that they were of a size that nevertheless inherently gave rise to suitable scattering effects or were fully or partially reflective.

Although the present invention has been described above in respect of its application to the formation of an organic layer comprising an array of pixels, it will be appreciated by the person skilled in the art that the method of the present invention can be used to form other patterned layers such as layers comprising one or more parallel rows or columns, each column or row comprising the same light-emissive material or comprising light-emissive materials which emit light of different colours. Alternatively, the method of the present invention can be used to form layers in the shape of one or more legends such as ones comprising one or more numerals or letters or other shapes, each numeral, letter or other shape composing the legend comprising the same light-emissive material or comprising light-emissive materials which emit light οf different colours.

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CLATMS

- 1. A method for forming a patterned layer of a light-emissive material on a substrate, comprising the steps of:
- providing a holed layer on the surface of the substrate, the holed layer being permanently attached to the substrate and defining a plurality of holes through which the underlying substrate is exposed; and
- applying a light-emissive material to the surface of the holed layer opposite the substrate and displacing the light-emissive material in fluid form across the surface of the holed layer so as to selectively deposit the material only in the holes of the holed layer.
- 2. A method according to claim 1 wherein the holed layer is provided by laminating a polymer layer with pre-formed holes to the surface of the substrate.
- 3. A method according to claim 1 wherein the holed layer is provided by providing a layer of a polymer on the substrate, and then forming the plurality of holes in the polymer layer.
- 4. A method according to claim 3 wherein the hoes are formed in the polymer layer by etching.
- 5. A method according to claim 4 wherein the etching is carried out using an oxygen-based plasma.
- 6. A method according to claim 4 or 5 wherein the etching is carried out by a plasma etching process.
- 7. A method according to claim 3 wherein the polymer layer is provided by coating the substrate with the polymer.

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- 8. A method according to claim 3 wherein the polymer layer is provided by laminating a pre-formed polymer layer to the substrate.
- 9. A method according to any preceding claim wherein the holed layer comprises an electrically insulating polymer.
- 10. A method according to claim 9 wherein the electrically insulating polymer is a fluorinated polymer.
- 11. A method according to any preceding claim wherein the thickness of the holed layer is in the range of 50 to 100nm.
- $12.\ \mbox{A}$  method for forming an optoelectronic device comprising the steps of:

providing a substrate having a patterned first electrode on a surface thereof;

providing a holed layer on the surface of the substrate having the first patterned electrode, the holed layer being permanently attached to the substrate and defining a first set of holes through which the pattered first electrode is exposed; applying a first light-emissive material to the surface of the holed layer opposite the substrate, and displacing the first light-emissive material in fluid form

across the surface of the holed layer so as to selectively deposit the material only in the first set of holes of the holed layer;

solidifying the first light-emissive material; and forming a second electrode on the solidified first light-emissive material in the first set of holes such that charge carriers can move between the first light-emissive material and the second electrode.

13. A method for forming an optoelectronic device according to claim 12 wherein the holed layer is provided by laminating a

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polymer layer with pre-formed holes to the surface of the substrate.

- 14. A method for forming an optoelectronic device according to claim 12 wherein the holed layer is provided by providing a layer of a polymer on the surface of the substrate having the patterned first electrode, and then forming the first set of holes in the polymer layer.
- 15. A method for forming an optoelectronic device according to claim 14 wherein the first set of holes are formed by etching.
- 16. A method for forming an optoelectronic device according to any one of claims 12 to 15 wherein the patterned first electrode comprises a two-dimensional array of discrete pixel electrodes.
- 17. A method according to claim 16 wherein the first set of holes only expose some of the discrete pixel electrodes.
- 18. A method according to claim 12 further comprising forming a second set of holes in the holed layer to expose discrete pixel electrodes which were not exposed by the first set of holes in the holed layer, depositing a layer of second light-emissive material in the second set of holes and forming a third electrode on the layers of second light-emissive material such that charge carriers can move between the layers of second light-emissive material and the third electrode.
- 19. A method according to claim 18 wherein the second electrode is additionally formed over substantially the entire surface of the holed layer opposite the substrate, and the second set of holes is formed by etching through the second electrode and the holed layer.

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20. A method according to claim 19 further comprising forming a third set of holes in the holed layer to expose further discrete pixel electrodes which were not exposed by the first or second set of holes, depositing a layer of third light-emissive material in the third set of holes, and forming a fourth electrode on the layers of third light-emissive material such that charge carriers can move between the fourth electrode and the layers of third light-emissive material.

- 21. A method according to claim 20 wherein the first, second and third light-emissive materials are each capable of emitting light of the three primary colours respectively.
- 22. A method according to any of claims 12 to 21 further comprising the step of forming a layer of a charge transport material in the first set of holes.
- 23. A method according to claim 22 wherein the layer of charge transport material is deposited in the first set of holes by applying the charge transport material to the surface of the holed layer and displacing the charge transport material in a fluid form across the surface of the holed layer whereby it is deposited in the first set of holes.
- 24. A method for forming an optoelectronic device according to any one of claims 12 to 23 wherein the second electrode is deposited without first removing the holed layer.
- 25. A method for forming an optoelectronic device according to claim 20 wherein the second, third and fourth electrodes are formed without removing the holed layer.
- 26. An optoelectronic device produced by the method of any of claims 14 to 25.







# (12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

### (19) World Intellectual Property Organization International Bureau



# 

#### (43) International Publication Date 14 December 2000 (14.12.2000)

(51) International Patent Classification7:

# PCT

# (10) International Publication Number WO 00/76008 A 1

(74) Agent: HARTWELL, Ian, Peter; Cambridge Display

- (21) International Application Number: PCT/GB00/01991
- (22) International Filing Date: 1 June 2000 (01.06,2000)
- (25) Filing Language:

English

H01L 51/20

(26) Publication Language:

English

GR

(30) Priority Data: 9913451.2 9913695.4

9 June 1999 (09 06 1999) GB 11 June 1999 (11.06.1999)

- (71) Applicant (for all designated States except US): CAM-BRIDGE DISPLAY TECHNOLOGY LIMITED [GB/GB]; Greenwich House, Madingley Rise, Madingley Road, Cambridge, Cambridgeshire CB3 0HJ (GB).
- (72) Inventors; and
- (75) Inventors/Applicants (for US only): BERGER, Paul, Raymond [US/US]; 17 West Ridge Court, Newark, DE 19711 (US), HEEKS, Stephen, Karl [GB/GB]; Cambridge Design Technology Limited, Madingley Rise, Madingley Road, Cambridge, Cambridgeshire CB3 0HJ

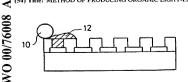
- Techology Limited, Greenwich House, Madingley Rise, Madingley Road, Cambridge, Cambridgeshire CB3 0HJ
- (81) Designated States (national): AE, AG, AL, AM, AT, AU, AZ. BA. BB. BG. BR. BY. CA. CH. CN. CR. CU, CZ. DE. DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR. HU. ID. IL., IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT LILLV MA. MD. MG. MK. MN. MW. MX, MZ, NO. NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.
- (84) Designated States (regional): ARIPO patent (GH. GM. KE, I.S. MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT. BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT. LU. MC. NL. PT. SE), OAPI patent (BF. BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

#### Published:

With international search report.

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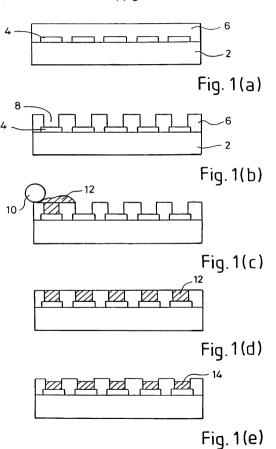
(54) Title: METHOD OF PRODUCING ORGANIC LIGHT-EMISSIVE DEVICES



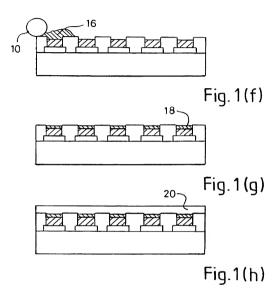
(57) Abstract: A method for forming a patterned layer of a light-emissive material on a substrate, comprising the steps of providing a holed layer on the surface of the substrate, the holed layer being permanently attached to the substrate and defining a plurality of holes through which the underlying substrate is exposed, and applying a light-emissive material to the surface of the holed layer opposite the substrate and displacing the light-emissive material in fluid form across the surface of the holed layer so as to selectively deposit the material only in the holes of the holed layer.

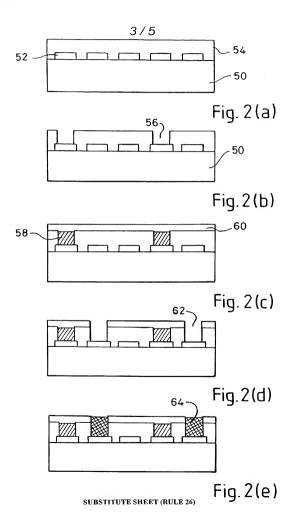


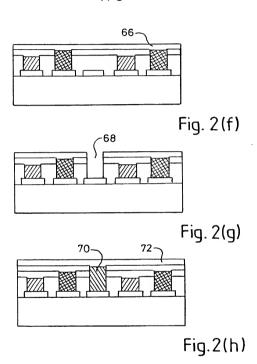
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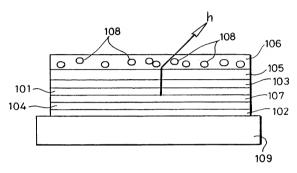


Fig. 3

# DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that my residence, post office address and citizenship are as stated below next to my name; I believe that I am an original, first and joint inventor of the subject matter which is claimed and for which a patent is sought on the invention entitled "METHOD OF PRODUCING ORGANIC LIGHT-EMISSIVE DEVICES," the specification of which was filed on June 1, 2000, amended on December 3, 2001, and assigned Application Serial No. 10/009,078. I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment(s) referred to above. I acknowledge the duty to disclose to the Patent and Trademark Office all information known to me to be material to patentability as defined in 37 C.F.R. §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

			Priority Claimed	
9913451.2	Great Britain	09 June 1999	Ø O	
(Application Serial Number)	(Country)	(Day/Month/Year Filed)	Yes No	
			Priority Claimed	
9913695,4	Great Britain	11 June 1999		
(Application Serial Number)	(Country)	(Day/Month/Year Filed)	Yes No	

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below:

(Application Serial Number) (Day/Month/Year Filed)

I hereby claim the benefit under 35 U.S.C. §120 of any United States application(s) or PCT international application(s) designating the United States of America listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior application(s) in the manner provided by the first paragraph of 35 U.S.C. §112, I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in 37 C.F.R. §1.56 which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:

 PCT/GB00/01991
 01 June 2000
 Pending

 (Application Serial Number)
 (Day/Month/Year Filed)
 (Status-Patented, Pending or Abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. §1001 and that such willful false statements may ieopardize the validity of the application or any patent issued thereon.

### APPLICABLE RULES AND STATUTES

# 37 CFR 1.56. DUTY OF DISCLOSURE - INFORMATION MATERIAL TO PATENTABILITY (Applicable Portion)

- (a) A patent by its very nature is affected with a public interest. The public interest is best served, and the most effective patent examination occurs when, at the time an application is being examined, the Office is aware of and evaluates the teachings of all information material to patentability. Each individual associated with the filing and prosecution of a patent application has a duty of candor and good faith in dealing with the Office, which includes a duty to disclose to the Office all information known to that individual to be material to patentability as defined in this section. The duty to disclose information exists with respect to each pending claim until the claim is canceled or withdrawn from consideration, or the application becomes abandoned. Information material to the patentability of a claim that is canceled or withdrawn from consideration need not be submitted if the information is not material to the patentability of any claim remaining under consideration in the application. There is no duty to submit information which is not material to the patentability of any existing claim. The duty to disclose all information known to be material to patentability of any claim remaining under consideration in the application known to be material to patentability of any claim formation which is not material to the patentability of any existing claim. The duty to disclose all information known to be material to patentability of any claim issued in a patent was cited by the Office or submitted to the Office in the manner prescribed by §§ 1.97(b)-(d) and 1.98. However, no patent will be granted on an application in connection with which fraud on the Office was practices or attempted or the duty of disclosure was violated through bad faith or intentional misconduct. The Office encourages applicants to carefully examine:
  - (1) prior art cited in search reports of a foreign patent office in a counterpart application, and
  - (2) the closest information over which individuals associated with the filing or prosecution of a patent application believe any pending claim patentability defines, to make sure that any material information contained therein is disclosed to the Office.

Information relating to the following factual situations enumerated in 35 USC 102 and 103 may be considered material under 37 CFR 1.56(a).

# 35 U.S.C. 102. CONDITIONS FOR PATENTABILITY: NOVELTY AND LOSS OF RIGHT TO PATENT

A person shall be entitled to a patent unless --

- (a) the invention was known or used by others in this country, or patented or described in a printed publication in this or a foreign country, before the invention thereof by the applicant for patent, or
- (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of the application for patent in the United States, or
  - (c) he has abandoned the invention, or
- (d) the invention was first patented or caused to be patented, or was the subject of an inventor's certificate, by the applicant or his legal representatives or assigns in a foreign country prior to the date of the application for patent in this country on an application for patent or inventor's certificate filed more than twelve months before the filing of the application in the United States, or
- (e) the invention was described in a patent granted on an application for patent by another filed in the United States before the invention thereof by the applicant for patent, or on an international application by another who has fulfilled the requirements of paragraph (1), (2), and (4) of section 371(c) of this title before the invention thereof by the applicant for patent, or
  - (f) he did not himself invent the subject matter sought to be patented, or
- (g) before the applicant's invention thereof the invention was made in this country by another who had not abandoned, suppressed, or concealed it. In determining priority of invention there shall be considered not only the respective dates of conception and reduction to practice of the invention, but also the reasonable diligence of one who was first to conceive and last to reduce to practice, from a time prior to conception by the other.

# 35 U.S.C. 103. CONDITIONS FOR PATENTABILITY; NON-OBVIOUS SUBJECT MATTER (Applicable Portion)

A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

Subject matter developed by another person, which qualifies as prior art only under subsection (f) or (g) of section 102 of this title, shall not preclude patentability under this section where the subject matter and the claimed invention were, at the time the invention was made, owned by the same person or subject to an obligation of assignment to the same person.

## 35 U.S.C. 112. SPECIFICATION (Applicable Portion)

The specification shall contain a written description of the invention, and of the manner and process of making and using it, in such full, clear, concise, and exact terms as to enable any person skilled in the art to which it pertains, or with which it is most nearly connected, to make and use the same, and shall set forth the best mode contemplated by the inventor of carrying out his invention.

POWER OF ATTORNEY: 1 hereby appoint as my attorneys, with full powers of substitution and revocation, to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

John B. Lungmus(1<u>8</u>,56<u>6</u>) Allen H. Gerstein (<u>22,218</u>) Nate F. Scarpelli (<u>22,320</u>) Michael F. Borun (<u>25,447</u>)-Carl E. Moore, Jr. (<u>26,487</u>) Richard H. Anderson (<u>26,526</u>) Patrick D. Ertel (26,877) Richard B. Hoffman(26,910) James P. Zeller (28,491) Kevin D. Hogg (31,839) Jeffrey S. Sharp (31,879), Martin J. Hirsch (32,237) James J Napoli (32,361) Richard M La Barge (32,254) Robert M. Gerstein (34,824) Anthony G. Sitko (36,278)-James A. Flight (37,622) Roger A. Heppermann (37,641)
David A Gass (38,153)
Gregory C Mayer (38,238)
Michael R. Weiner (38,359)
William K. Merkel (40,728)
Andrew M. Lawrence (46,130)

	Send correspondence to:	James P. Zeller					
	FIRM NAME	PHONE NO.	STREE	et e	CITY & STATE	ZIP CODE	
	Marshall, Gerstein & Borun 312-474-63		6300 Sears Tower 233 South Wacker Drive		Chicago, Illinois	60606-6357	
الاء	Full Name of First or Sole Inventor			Citizenship			
SUI	Paul R. Berger_			United States of America			
	Residence Address - Street			Post Office Address - Street			
- 1	4290 Mountview Road			4290 Mountview Road			
	City (Zip) Columbus 43220			City (Zip) Columbus 43220			
				State or Country			
1	State or Country Ohio			Ohio			
	Date May 30,	2002		Signature (	Jul By		
ı	7				//		
0	Second Joint Inventor, if any			Citizenship			
U	Stephen Karl Heeks			Great Britain			
1	Residence Address - Street		l	Post Office Addr			
- 1	127 Rampton Road			127 Rampton	Road		
	City (Zip)			City (Zip) Cottenham, Cambridgeshire CB4 8TJ			
	City (Zip) Cottenham, Cambridgeshire CB4 8TJ GDX State or Country			State or Country			
	United Kingdom			United Kingdom			
	Date 5/6/200 Z			Signature & Hells			